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THE APPLICATION OF THERMAL PLASMA TO EXTRACTION  
METALLURGY AND RELATED FIELDS

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14. Abstract Various applications of thermal plasma to extraction metallurgy and related fields are surveyed, chiefly on the basis of documents published during the past two or three years. Applications to melting and smelting, to thermal decomposition, to reduction, to manufacturing of inorganic compounds, and to other fields are considered.			15. Type of Report and Period Covered Translation		
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# THE APPLICATION OF THERMAL PLASMA TO EXTRACTION METALLURGY AND RELATED FIELDS

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## 1. Introduction

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More than 20 years have already gone by since plasma torches, which generate thermal plasma flow, were developed and research was started on their applications to high-temperature metallurgical reactions. Two types of torches have been established at the present time. One is the type which utilizes D.C. or A.C. arc discharges between the electrodes, and the other is the type which utilizes high-frequency induction discharges with no electrodes. They may be said to be the best heat sources of ultra-high temperatures, but they have a relatively limited record of applications and practical use in the past in the fields of extraction metallurgy and synthesis of inorganic compounds. Various reactors have been trial-manufactured for each specific purpose, and tests have been carried out repeatedly. However, in most cases they have remained at the research stage, and the distinctive features of plasma have not been utilized to the full.

In manufacturing processes utilizing plasma, naturally, the points listed below must be taken into consideration. 1) The plasma gases must have a high temperature and reactivity. 2) The investment in equipment and the raw material costs must be low, and the products must have a high added value. 3) Remarkable improvements or special changes must be brought about in the physical and chemical properties of the products, and these must coincide with the specific purposes for which they are to be used. 4) The multi-stage processes must be simplified. 5) The processes must cause no environmental pollution, must lead to economy of natural resources, and must lend themselves to effective utilization of

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\* Numbers in the margin indicate pagination in the foreign text.

the waste heat. 6) There must be a secure market for the products which displays promise of future development. In addition, there are many who believe that the goal for the time being ought to be a process which can be fully justified economically even when the electric power consumption for one reactor or reactor furnace will be 1 MW or less, which would be considerably less than that in the arc smelting furnaces of the past.

The past studies concerning applications of thermal plasma are outlined in the references given at the end of this paper, especially in the explanatory and general documents [1-21]. Therefore, we shall confine ourselves here to describing only the most recent trends in research. The items to be dealt with here are the following: comparisons of various types of plasma torches; melting and smelting; thermal decomposition; reduction; and others (synthesis of inorganic compounds).



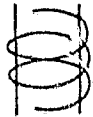
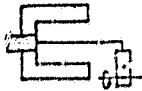
## 2. Comparisons of Various Types of Thermal Plasma Generating Devices

According to the results of the investigation by C.D. Schnell et al. [22], they can be summed up as shown in Table 1.

## 3. Melting and Smelting

This is a good example of an application which has reached the stage of industrialization. There are recent published explanations by Sugiyama et al. [23] and Ono [24]. The basic shape of the melting furnace is that of the Union Carbide company [25], but development has also made headway in Japan, the U.S.S.R., East Germany and Czechoslovakia. The plasma torch consists of a cathode (usually made of tungsten with thorium) and a water-cooled copper nozzle. The metal to be melted is used as the anode. It is bombarded directly with a plasma arc of an inert gas, and the heat is transferred efficiently from the plasma to the metal. This primary melting by means of a plasma arc is called PAM (an abbreviation for Plasma Arc Melting), and the remelting of the casting after the primary melting is called PAR (an abbreviation for Plasma Arc Remelting). Ordinarily in PAM a melting containment equipped with a refractory lining is tilted over to pour out the molten metal to form ingots, and in PAR

TABLE 1.  
COMPARISONS OF VARIOUS TYPES OF THERMAL PLASMA GENERATING DEVICES

1 形 式	2 直 流 アー ク	3 三 相 交 流 アー ク	4 高 周 波 アー ク	5 液 体・安 定 化* 直 流 アー ク
				
6 通常の工業用トーチの能力	12 150~200 kW** (カーボンアーク) (では 1MW 以上)	13 500~1000 kW (カーボンアーク) (では 1MW 以上)	100~1000 kW	300~500 kW
7 kW 当りの設備投資 (200 kW の装置)	280 \$/kW	330 \$/kW	450 kHz 700~1000 \$/kW 10~11 Hz 200~250 \$/kW	300 \$/kW
8 主たる使用ガスまたは液体	14 貴ガス, 窒素, 水素, 炭化水素	15 貴ガス, 窒素, 水素, 炭化水素	16 とくに制約なし	17 絶 縁 性 液 体 (水, アルコール, 炭化水素など)
9 プラズマの化学的 特性	18 中性, 還元性	19 中性, 還元性	20 中性, 還元性, 酸化性	21 中性, 還元性, 酸化性
10 トーチの効率	60~80%	~80%	~50%	85~90%
11 電源効率	90~95%	95%	30~80%	90~95%

\* Liquid-stabilized d.c. arc のこと, アークの周囲を液流で囲んで安定化させる。 22

\*\* 金属の溶解炉としては 1 MW 以上のものがある。 23

- Key: 1. Type  
2. D.C. arc  
3. Three-phase A.C. arc  
4. High-frequency arc  
5. Liquid-stabilized D.C. arc  
6. Capacity of ordinary torches for industrial use  
7. Plant investment per kW (200 kW equipment)  
8. Main gases or liquids used  
9. Chemical characteristics of the plasma  
10. Efficiency of the torch  
11. Power source efficiency  
12. 150-200 kW\*\* (with carbon arc, 1 MW or more)  
13. 500-1000 kW (with carbon arc, 1 MW or more)  
14. Rare gases, nitrogen, hydrogen, hydrocarbons  
15. Rare gases, nitrogen, hydrogen, hydrocarbons  
16. No particular restrictions  
17. Liquids with insulating properties (water, alcohol, hydrocarbons, etc.)  
18. Neutral, reducing  
19. Neutral, reducing  
20. Neutral, reducing, oxidizing  
21. Neutral, reducing, oxidizing  
22. \*Liquid-stabilized D.C. arc. The arc is surrounded by a liquid current and stabilized.  
23. \*\*There are furnaces for melting metals which have a capacity of 1 MW or more.

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ingots are formed continuously by means of the water-cooled copper mold.

In our country there has been developed a plasma induction melting /343 (PIM) method, in which, in addition to plasma arc heating, induction heating and stirring of the molten metal are also performed [26]. The maximum rated current of the plasma torch is 3,000 A (voltage: 150 V). The maximum electric power applied is about 1 MW; this is the total of the arc heating, induction heating and stirring. Melting of 2 tons of stainless steel is possible. The largest furnace of this type in the world is in operation in East Germany at the VEB Edelstahlwerk Freital [27, 28]. Four torches are installed on the top of the side walls of the furnace. The ratings and basic units are shown in Table 2.

TABLE 2.  
RATINGS AND BASIC UNITS OF EAST GERMAN PLASMA MELTING FURNACE [28]

1 トーチ (4 本) の全出力	12~15 MW	4 エネルギー原単位 (溶解期)	625 kWh/t
2 アーク電圧 (1 本のトーチ)	150~610 V	5 アルゴンガス原単位	60 m <sup>3</sup> /h
3 最大アーク電流 (1 本のトーチ)	10,000 A	6 冷却水原単位	60 m <sup>3</sup> /h

Key: 1. Overall output of 4 torches  
2. Arc voltage (one torch)  
3. Maximum arc current (one torch)  
4. Energy basic unit (melting period)  
5. Argon gas basic unit  
6. Cooling water basic unit

One might list the following advantages of PAM and PIM. 1) The plasma arc has stability and good heat conduction. 2) Deoxidation and decarburization are performed forcibly by melting in the presence of an inert gas at atmospheric pressure. 3) Deoxidation and desulfurization are performed effectively by utilizing slag (slag smelting). 4) There is a good yield of the alloy components. 5) There is a reduction of the amount of inclusions. 6) There is the possibility of special refining by means of reactive gases such as hydrogen, nitrogen, etc.

The problems are the following. 1) The speed of removal of the impurities. 2) The useful life of the plasma torches. 3) The service

life of the refractories in the furnace body. 4) The price of the argon gas. 5) The electric power basic unit. The above-mentioned PIM method has produced good results in the melting of extremely low-carbon stainless steels, sealed alloys of the iron-nickel-cobalt system, magnetic alloys of the iron-nickel-manganese (plus molybdenum) system, copper alloys for electrically conductive materials, heat-resistant alloys AISI 1660, etc. [26, 29, 30].

Research on PAR is being conducted especially energetically in the Soviet Union, where it appears that a 2,000 kW furnace with 6 plasma torches for manufacturing 5-ton steel ingots is being tested [31]. In East Germany as well, a number of 1,200 kW furnaces have been built [32]. In our country, 150 kW furnaces have been developed [22, 23, 32], but appended to them are water-cooled molds which have built-in coils for generating induction fields for stirring the molten metal. In this system, the base metal for remelting is supplied in an obliquely downward direction to the vertical plasma torch, and the ingots are drawn down continuously from the molds.

One may mention the following characteristic features of these plasma remelting methods. 1) Just as in the electron beam remelting method, the plasma output and the remelting speeds can both be controlled independently. 2) The remelted ingots have a good surface texture (close to that of electroslag melting), and there is a high working yield. 3) It is easy to separate inclusions and cause them to float up. 4) The molten metal pool is shallow, and a texture close to that of one-directional solidification can be obtained. 5) It is possible to manufacture ingots with irregularly shaped cross sections. 6) Slag refining and special refining by means of reactive gases are possible. 7) Not only bar-shaped raw materials, but also raw materials with granular or flake shapes can also be charged. /344

Remelting of various types of steels and superalloys, as well as of titanium, chromium and chromium alloys has been attempted [31-35], but these methods ought to be called methods still at the stage of research and development. Research has been under way for some time in the Soviet



Union [36] about stainless steel in which supersaturated nitrogen is put in solid solution in the molten steel thanks to the nitriding effects from nitrogen plasma. There are very interesting reports [37-40] about fundamental research concerning the denitriding or decarburizing mechanisms of molten steel in the presence of hydrogen. It is also anticipated that there will be developments in research on slag-metal reactions during plasma arc melting [41]. Attention is focused on the melting of ferrite type stainless steels with extremely low carbon, nitrogen and sulfur concentrations obtained by using flux for desulfurization [42].

Since the plasma arcs used in the above-mentioned plasma melting are restricted to arc columns which have a narrow high-temperature region, they are not suited for rapid heating of large areas. For this reason, methods for using A.C. magnetic fields to oscillate the plasma arcs and increase their widths are being studied [43]. There have also been developed large-size plasma torches which have expanded reaction areas and in which active oxygen is mixed with the argon to enable them to withstand use over prolonged periods [44].

The plasma electron beam method (HHCD method, an abbreviation for Hot Hollow Cathode Discharge) is among the methods which have attracted attention for melting and refining metals at a vacuum of medium degree (several Torr to  $10^{-4}$  Torr), unlike the PAM and PAR methods, which are ordinarily implemented at normal pressures. An electron current is extracted from plasma formed by glow discharge of an inert gas inside a hollow cathode made of tantalum; the current is focused and bombarded on the anode which is to be heated. 3-ton ingots have been prepared in titanium melting [45, 46].

In the Soviet Union success has also been achieved [47] in growing large-size single crystals (50 mm in diameter) of high-melting-point metals (tungsten, molybdenum, rhenium, niobium) and high-melting-point compounds (carbides of tantalum and hafnium) by utilizing plasma melting.

Arc plasma has been utilized for a long time for machining purposes such as welding and fuse melting, but another important application is

to spray coating, in which novel developments are anticipated. Consideration of this will be omitted here because it is somewhat removed from the field of extraction metallurgy. The reader is referred to suitable explanations in the literature [48-53].

Research has also been under way for a long time on methods by which powders of metals, alloys and compounds of a suitable particle size are melted at a high plasma temperature and spheroidized by their surface tension [54-57]. There has been a very recent report about spheroidizing of aluminum silicate particles by means of a three-phase A.C. plasma furnace [58].

#### 4. Thermal Decomposition

Considerable experience has already been accumulated concerning the method of using three-phase plasma arc furnaces of the MW class to produce thermal decomposition of methane or liquid hydrocarbons and manufacture acetylene or ethylene [59]. However, this method has not developed in view of problems concerning the electric power basic units and market difficulties.

A famous example in which this was successfully applied to powdered ores is the process by which zirconium sand (chief component  $ZrSiO_4$ ) is thermally decomposed into zirconium oxide and silicon oxide [60, 61]. The type of the furnace is one in which three consumable graphite anodes are arranged at  $120^\circ$  intervals below one plasma torch (tungsten cathode). Argon, nitrogen or air arcs with a large volume are formed by discharges between them, and powdered ores of 50  $\mu m$  or less are introduced from the periphery very close to the cathode. This method is considered to have a high efficiency and has also been proven theoretically [62, 63]. There is another method in which three plasma torches are used instead of the three consumable anodes [64]. The electric power basic unit is around 2 kWh per kg of raw material.

Thermal decomposition of molybdenite shipping ore (main component  $MoS_2$ ) has been tested in Canada [65, 66]. However, when an attempt was

made to attain a high decomposition rate, the electric power basic unit increased. Therefore, the orientation was changed towards developing a process for manufacturing ferromolybdenite using a furnace of the above-mentioned PAR system in which molybdenum would be alloyed with the molten iron of the anode simultaneously with thermal decomposition. Kinetic research has also been performed concerning the thermal decomposition reaction [67]. In some of the research, the experiments were carried out more than a decade ago, and the results have only recently been made public [68]. Important research topics include the elucidation of the path of movement of the powder particles introduced inside the plasma, the variations in their speed, their time of residence, the movement of the heat from the plasma, and the physical and chemical changes accompanying it. A number of fundamental studies have been made /345 on these subjects [69-74].

## 5. Reduction

When Nakamura et al. [75] melted and reduced iron ore by means of hydrogen plasma arcs, they obtained molten iron of a high purity, the removal of phosphorus by volatilization was remarkable, and the rate of utilization of hydrogen in reduction was considerably higher than the theoretical value. It is considered that a role may be played in the reaction by atomic hydrogen. Gold and MacRae [76, 77] developed devices with a maximum output of 1 MW in which the plasma torches themselves were used as the reactors. The molten ore forms a film on the inner wall of the torch and protects it, while the heat of the plasma is absorbed, the reaction takes place, and the ore drops down into the crucible below, where it is further heated by the plasma. Natural gas and hydrogen were charged in together with the powdered ore and were reduced. However, both the electric power basic unit and the reduction gas basic unit diverge considerably from the theoretical values. Other experiments have been conducted with similar devices in which vanadium trioxide obtained by preliminary reduction of vanadium pentoxide was supplied together with coke powder, the reduced vanadium was alloyed with the molten iron in the crucible, and ferrovanadium was produced. The economy of this method has been proven [78]. In France as well,

a method of direct iron-making from iron ores by means of plasmas of hydrogen and natural gas has been tested on a scale of 1 MW. The method has been found to be profitable thanks to the increased furnace volume and the utilization of the waste gases, but only the bare details have been made public [79].

When ilmenite (main component  $\text{FeO} \cdot \text{TiO}_2$ ) is melted and reduced by means of hydrogen plasma, it can be separated easily into iron and a slag which has a high concentration of titanium oxide [80]. The efficiency of utilization of the hydrogen is high, and there is extremely good coagulation and growth of the iron. When methane plasma is used, the active carbon produced by decomposition plays the predominant role in reduction, and the waste gases are high-temperature hydrogen and carbon monoxide [81]. A recent report of Chase et al. [82] mentions that a method has been adopted in which both methane gas and powdered ilmenite ore are supplied into a graphite cylinder in which hydrogen and argon plasma is formed. Although the details of the construction of the reactor are not published, they assert that the method has a high economy.

Another reaction furnace has been developed in which the powdered ore is supplied in the opposite direction to the flow of the high-temperature exhaust gas produced by the reaction, and the ore is heated and given preliminary reduction. Then the ore is dropped down into a three-phase A.C. plasma arc formed while passing the reduction gas from three hollow graphite electrodes, and the product is kept in a molten state at the bottom of the arc [83, 84]. Experiments have also been carried out in which iron-chromium alloys were manufactured using a low-grade powdered chromite ore and coal as the reducing agent [85]. Fey et al. [86] have also announced plans for a 16 MW methane plasma reducing plant for chromite. It is thought that there are many who would doubt the economy in connection with the electric power basic units.

There are reports [87-90] of plasma thermal reduction methods using carbon for various types of oxides such as niobium pentoxide, vanadium pentoxide, silicon oxide and aluminum oxide, but almost all of them have

not gone beyond the stage of small-scale experiments. In the research of Tylko [90], a special reaction furnace is used, in which the tip of the cathode rotates along the anode, which has the shape of a round ring, the arc also rotates at the same time, and a large plasma volume is formed. Govo et al. [91] have studied the possibility of collecting metals by thermal decomposition of oxides.

Recent studies on the reduction of halides [92-95] have dealt with thermal reduction of boron trichloride, titanium tetrachloride and silicon tetrachloride in hydrogen plasma. Methods of depositing amorphous silicon from silicon tetrachloride and hydrogen during glow discharge at a low temperature and pressure, rather than at normal pressure and high temperature, have begun to be studied along with methods departing from silane [96]. Studies are also being made of the mass production of inexpensive polycrystalline silicon for use in solar batteries. Pilot studies have been carried out in which silicon tetrachloride and sodium as the reducing agent were supplied in a spray state into a mixed gas consisting of arc-heated argon and hydrogen [97]. While silicon is produced continuously, sodium is recovered at the same time from the NaCl produced in this way. Progress is also being made with similar experiments using equipment in which a D.C. plasma torch is combined with a silicon carbide reaction tube [98].

In England work is now underway to develop a process by which iron powder or iron alloy powder is manufactured by hydrogen reduction from iron chlorides or mixtures of them and chlorides of alloy elements [99, 100]. A characteristic feature is the fact that plasma is used as the heating source for the hydrogen.

There is another method in which a vertical rotary furnace is used, the admixtures are pressed against the furnace walls by centrifugal force, they are dissolved by the heat of the plasma, and they are made to flow down while forming a film. Experiments have been performed in which the tin contained in slag was evaporated and recovered as an oxide (SnO) [101].

## 6. Other: Manufacturing of Inorganic Compounds, etc.

Research is going forward in methods in which silicon tetrachloride is introduced into high-frequency induced oxygen plasma in order to /346 synthesize quartz ingots of an ultra-high purity which will serve as the materials for glass fibers for the transmission of light [102]. It seems that the stage has now been reached where industrial production is possible at any time in response to an increase in demand. There is also being developed a method for adding specific elements as impurities in suitable concentrations in order to vary the refractive index. It will be interesting to watch the competition with the method of utilizing reactions in the presence of depressurized, unbalanced plasma, such as microwave discharge [102], and the non-plasma method. Titanium oxide for pigments is apparently being manufactured in the Soviet Union from titanium tetrachloride by a similar system [7], but the details are unclear.

It is possible to synthesize fine powders of nitrides or carbides (for example, silicon carbide or silicon nitride) by a combination of plasma heating and quenching, starting out from halides (chiefly chlorides) and nitrogen, hydrogen (or ammonia) or hydrocarbon gas [94, 104, 105]. There is also a method for evaporating metal powders in a plasma and forming compounds of them, ultra-fine particles of them, or fine alloy particles by condensation [106]. Research on plasma "CVD" is also coming to be highly active. In this method, the plasma is used under a reduced pressure, and metals and their compounds are deposited as films on the surface of a suitable material, rather than as fine powders. The reaction proceeds at a lower temperature than in the conventional methods of the past, and the characteristic features of the deposited substances are different. These and other distinctive features are noted in cases where a thermally unbalanced plasma is utilized.

The reader should consult general articles [14-16, 107] concerning the plasma synthesis of nitrides and carbides from oxides.

## 7. Conclusion

We have surveyed research in applications of plasma to extraction metallurgy and the related fields, chiefly on the basis of documents which have been published during the past two or three years. Many studies have been omitted from the survey, but the author will be pleased if the reader has been able to understand the overall trends. We touched to a certain degree upon the applications of low-pressure, low-temperature plasma as well. The author begs forgiveness if this has overlapped to some extent with other articles.

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\* Numbering mistake in original. - Tr.